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13. ABSTRACT *(maximum 200 words)*

Accomplishment of researches conducted under support of contract DAAL03-89-K-0119 have been reported. Two Ph. D. degrees have been granted to two students supported by this contract. A total of six categories of studies, designated for convenience, have been performed. The first five categories of studies concern the point defects and diffusion mechanisms of elements occupying the group III sublattice of GaAs and/or AlAs/GaAs superlattice materials, and one category of studies concerns the group V sublattice acceptor carbon in AlAs/GaAs superlattices. With theses studies, our knowledge of point defects and diffusion mechanisms of elements occupying the group III sublattice of GaAs and/or AlAs/GaAs superlattice materials have now becoming substantially complete in the sense that interpretations of almost all experimental results are self-consistent and mutually-consistent.

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INVESTIGATION OF SUPERLATTICE DISORDERING AND DIFFUSION MECHANISMS IN GaAs

FINAL REPORT

T. Y. Tan and U. M. Gösele

US ARMY RESEARCH OFFICE
CONTRACT
DAAL03-89-K-0119

DEPARTMENT OF MECHANICAL ENGINEERING
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ABSTRACT

Accomplishment of researches conducted under support of contract DAAL03-89-K-0119 have been reported. Two Ph. D. degrees have been granted to two students supported by this contract. A total of six categories of studies, designated for convenience, have been performed. The first five categories of studies concern the point defects and diffusion mechanisms of elements occupying the group III sublattice of GaAs and/or AlAs/GaAs superlattice materials, and one category of studies concerns the group V sublattice acceptor carbon in AlAs/GaAs superlattices. With theses studies, our knowledge of point defects and diffusion mechanisms of elements occupying the group III sublattice of GaAs and/or AlAs/GaAs superlattice materials have now becoming substantially complete in the sense that interpretations of almost all experimental results are self-consistent and mutually-consistent.

I. PROJECT SUMMARY

The research program supported by ARO contract DAAL03-89-K-0119 is entitled *INVESTIGATION OF SUPERLATTICE DISORDERING AND DIFFUSION MECHANISMS IN GaAs.* The principal investigators are T. Y. Tan, U. M. Gösele and R. Tsu. The performance period was from 8/1/89 to 3/31/94, including two no-cost extension periods. The first was from 2/1/92 to 12/31/92, for the purpose of effectively utilizing resources then available for accomplishing more works. The second was from 7/1/93 to 3/31/94, for the purpose of managing the return of the leased ^{69}Ga and ^{71}Ga isotopes to Oak Ridge National Laboratory. In the second extension period, no further research work has been conducted under the support of the contract.

The purpose of conducting the studies supported by this project was to obtain consistent interpretations of existing experimental results, as well as to conduct the needed new experiments, concerning the point defect an diffusion phenomena occurring to elements occupying the group III sublattice in GaAs and in AlAs/GaAs superlattice materials, which are relevant to device fabrications. Previously, such a consistent interpretation did not exist. As a result of the present studies, our knowledge on the nature of point defects and diffusion mechanisms on the Ga sublattice of GaAs has become substantially systematic during the last few years. This means that interpretations of different experimental results are now becoming self- and mutually consistent.

II. Ph. D. STUDENTS

Under support of this contract, two students have received their Ph. D. degrees from the Department of Mechanical Engineering and Materials Science, Duke University. The first is Dr. Shaofeng Yu (1991) who is currently employed by Intel Corporation, Portland, OR. Dr. Yu's dissertation title is *STUDIES OF POINT DEFECT EQUILIBRIA AND IMPURITY DIFFUSION MECHANISMS IN GALLIUM ARSENIDE.* The second is Dr. Horng-Ming You (1993) who is currently employed by Advanced Micron Device Corporation, San Jose, CA. Dr. You's dissertation title is *SELF-DIFFUSION AND DIFFUSION OF SILICON, CARBON AND TRIPLY-NEGATIVELY-CHARGED Ga VACANCIES IN GaAs AND AlAs/GaAs SUPERLATTICES.*

III. TECHNICAL ACCOMPLISHMENT

Under support of this contract, point defects and diffusion mechanisms governing elements occupying the group III sublattice in GaAs and related materials, e.g., AlAs/GaAs superlattices, have been studied. These include the following subjects: (i) the diffusion mechanism and effects of the p-type dopants Zn and Be; (ii) the diffusion mechanism of the deep donor Cr; (iii) the diffusion mechanism and effects of the n-type dopant Si; (iv) the group III sublattice point defect properties in GaAs and related materials; (v) a ^{69}GaAs/^{71}GaAs superlattice (SL) disordering experiment. Prior to these studies, a consistent interpretation of point defects and the diffusion mechanisms in GaAs concerning the Ga sublattice did not exist. As a result of these studies, our knowledge on the nature of point defects and diffusion mechanisms on the Ga sublattice of GaAs has presently become substantially systematic, i.e., that interpretations of different experimental results are now becoming self- and mutually consistent. Most of the knowledge gained from the present studies have been accepted by the scientific community by now. In addition, we have also conducted a preliminary study of the diffusion mechanism and effects of the p-type dopant C occupying group V sublattice.

In the following, summaries of the above mentioned studies are presented.

III.1. Diffusion and effects of the p-type dopants Zn and Be

Existing experimental results in the literature have been first analyzed to study the Zn diffusion mechanism and its effect on AlAs/GaAs SL disordering.[1,2] It is concluded that Zn diffusion (inferred also for Be) is governed by the kick-out mechanism of the interstitial-substitutional impurities. The point defect species involved in this kick-out mechanism is the doubly-positively-charged group III or Ga sublattice self-interstitial, I_{Ga}^{2+}. By fitting Zn indiffusion profiles, the I_{Ga}^{2+} contribution to Ga self-diffusivity value in GaAs, D_{Ga}^{I}, has been obtained at 1100°C.[1] This is one factor allowed the determination of D_{Ga}^{I} values as a function of experimental temperature, discussed in section III.3. Using such D_{Ga}^{I} values, a simulation has been performed on the effect of Zn indiffusion on the AlAs/GaAs SL disordering.[2] Fits were obtained between the simulation results and some available experimental results.[2] In collaboration with a German group, experiments were performed to study extended defect formation phenomena induced by Zn indiffusion. Dislocations and voids containing Ga-precipitates were found.[3] The formation mechanism of these defects has been proposed.[3,4] Two

most important findings[3] are that (a) in almost all experiments conducted in the past, the Zn source is not a thermal equilibrium one, which leads to severe transient changes in the Zn indiffusion process, the GaAs crystal composition, and the governing point defects (I_{Ga}^{2+}) concentrations; and (b) irrespective of the Zn source condition, in experiments invoking an As-rich ambient, the GaAs crystal will never attain a one composition condition. This is because Zn tends to produce a Ga-rich material and is obtained at the diffusion front portion of the Zn profile, while the As-rich ambient renders the GaAs crystal also As-rich at its surface portion. The common practice of using an As-rich ambient was for the purpose of ensuring the Zn diffused region of the GaAs crystal in a single As-rich composition. This premise had not been fulfilled in any experiment.

III.2. Diffusion mechanism of the deep donor Cr

The deep donor Cr is also an interstitial-substitutional impurity occupying the Ga sublattice. Analyzing existing experimental results, it is found that (a) under indiffusion conditions, Cr diffuses via the kick-out mechanisms for which the governing point defect species is I_{Ga}^{2+}; and (b) under outdiffusion conditions, Cr diffuses via the dissociative mechanisms for which the governing point defect species is the triply-negatively-charged Ga vacancies, V_{Ga}^{3-}. As a results, the Ga self-diffusivity values contributed by I_{Ga}^{2+} (D_{Ga}^{I}), and contributed by V_{Ga}^{3-} (D_{Ga}^{V}) have both been extracted from fitting available Cr diffusion profiles.[5]

III.3. Group III sublattice point defect properties in GaAs and related materials

Using results obtained in our previous and present studies,[1,4-6] an estimate of D_{Ga}^{V} and D_{Ga}^{I} values has been obtained.[7] They are given by

$$D_{Ga}^{V}(n_i,\text{1-atm}) = 2.9\text{x}10^{8} \exp(-6\ \text{eV}/k_B T)\ \text{cm}^2\text{s}^{-1}\ , \qquad (1)$$

$$D_{Ga}^{V}(n_i,\text{Ga-rich}) = 3.93\text{x}10^{12} \exp(-7.34\ \text{eV}/k_B T)\ \text{cm}^2\text{s}^{-1}\ , \qquad (2)$$

$$D_{Ga}^{I}(n_i,\text{1-atm}) = 6.05 \exp(-4.71\ \text{eV}/k_B T)\ \text{cm}^2\text{s}^{-1}\ , \qquad (3)$$

$$D_{Ga}^{I}(n_i,\text{Ga-rich}) = 4.46\text{x}10^{-4} \exp(-3.37\ \text{eV}/k_B T)\ \text{cm}^2\text{s}^{-1}\ . \qquad (4)$$

In these equations n_i refers to the intrinsic condition, 1-atm refers to the GaAs crystal being in equilibrium with an ambient with an As_4 pressure value equals to 1-atm,

Ga-rich refers to a GaAs crystal with its composition being at the thermodynamically allowed Ga-rich limit, k_B is Boltzmann's constant, and T is the absolute temperature. The values expressed by Eqs. (1)-(4) are by now fairly generally used in the literature, for two reasons. First, they are the only set of such values available. Second, they seem to be fairly accurate.

We have obtained thermodynamically correct expressions for the thermal equilibrium concentrations of the six single point defect species in GaAs (the two vacancies V_{Ga} and V_{As}, the two self-interstitials I_{Ga} and I_{As}, and the two anti-site defects As_{Ga} and Ga_{As}), including neutral and all charged species.[8,9] Presently, the needed thermodynamic parameters for performing numerical calculations are available only for the V_{Ga} species and the neutral antisite defect As_{Ga}. Therefore, only the thermal equilibrium concentrations of these species have been calculated.[8,9] An extremely important and interesting finding is that the V_{Ga}^{3-} thermal equilibrium concentration $C_{V_{Ga}^{3-}}^{eq}$ is temperature *independent* or exhibiting a small *negative* temperature dependence for highly n-doped materials, in the sense that when T is lowered $C_{V_{Ga}^{3-}}^{eq}$ is either unchanged or increased. The $C_{V_{Ga}^{3-}}^{eq}$ values exhibit the normal positive temperature dependence in that when T is lowered $C_{V_{Ga}^{3-}}^{eq}$ is decreased. This is a result of the fact that, the free energy released by the electrons are sufficient to completely compensate the *configurational* enthalpy for forming V_{Ga}^{3-}. There are several sets of experimental results requiring the assumption that the $C_{V_{Ga}^{3-}}^{eq}(n)$ values is attained at the onset of an experiment or that non-equilibrium points defects are generated during the experiment, which can only be satisfactorily explained by the temperature independent or negative temperature dependence of the $C_{V_{Ga}^{3-}}^{eq}(n)$ values. The alternative possibility that the $C_{V_{Ga}^{3-}}^{eq}(n)$ values are reached in an initial small time period after the onset of an experiment is not feasible. To address this last point, a simulation study has been performed and the results confirmed the *impossibility* of populating V_{Ga}^{3-} to the appropriate $C_{V_{Ga}^{3-}}^{eq}(n)$ value in a short time after the onset of experiments.[10] In order to perform this simulation, we have formulated a *diffusion-segregation* equation which is generally applicable to all problems for which the diffusion and segregation behaviors of a species need to be accounted for together.[10] This is the first time a general diffusion-segregation equation has become available. Task A of the present proposal is based on these studies.

III.4. Diffusion mechanism of the group III sublattice n-type dopant Si

In this study, a set of Si outdiffusion experiments using pre-doped GaAs wafers have been conducted.[6] The experimental results confirm our previously suggested diffusion mechanism of Si in GaAs: that the V_{Ga}^{3-} species are involved.[4] At the same temperature, however, the Si outdiffusion diffusivity values are several orders of magnitude larger than those obtained under Si indiffusion conditions.[4] This is explained by noticing the difference of the V_{Ga}^{3-} thermal equilibrium concentrations $C_{V_{Ga}^{3-}}^{eq}$ in the two types of experiments. Because $C_{V_{Ga}^{3-}}^{eq}$ is proportional to n^3, initially the GaAs crystal $C_{V_{Ga}^{3-}}^{eq}$ value is larger by many orders of magnitude in the outdiffusion experiments than in the indiffusion experiments. During the experiments, this trend maintains by the development of a V_{Ga}^{3-} supersaturation in the Si outdiffusion cases and a V_{Ga}^{3-} undersaturation in the Si indiffusion cases.

III.5. The ^{69}GaAs/^{71}GaAs SL disordering experiments

The ^{69}GaAs/^{71}GaAs SL disordering experiment had been originally designed to obtain Ga self-diffusivity values in the temperature range of around ~900°C so as to compare with those expressed by Eq. (1), which are complied from Ga self-diffusivity values at T>1100°C and Al-Ga interdiffusivity values at T<950°C. In the literature, there are two sets of Al-Ga interdiffusivity data which are characterized by a 4 eV activation enthalpy and are higher than those given by Eq. (1), which consist of the majority of the Al-Ga interdiffusivity data. In the present ^{69}GaAs/^{71}GaAs SL disordering experiments, the measured Ga self-diffusivity values from 850 to 960°C are in agreement with the two sets 4 eV Al-Ga interdiffusivity data but not with the majority of the data represented by Eq. (1).[11] Our ^{69}GaAs/^{71}GaAs SL structures, grown on Si-doped GaAs substrates, were undoped. After the disordering annealing, the substrate Si doping level has decreased due to Si outdiffusion into the immediate adjacent SL layers. The SL regions further away from the substrate where the Al-Ga interdiffusion rates were measured, however, remained intrinsic. The decrease of the substrate doping leads to a decreased substrate $C_{V_{Ga}^{3-}}^{eq}(n)$ value, and hence some V_{Ga}^{3-} must have been released. These released V_{Ga}^{3-} migrate into the still intrinsic SL regions, leading to a V_{Ga}^{3-} supersaturation. Thus, the present 4 eV Ga self-diffusion data is due to the V_{Ga}^{3-} supersaturation effects. The two sets of Al-Ga interdiffusion data which showed the 4 eV

activation enthalpy were obtained from undoped AlAs/GaAs SL grown also on n-type GaAs substrates doped by Si.

III.6. Diffusion mechanism of the p-type dopant C

In this study, a set of experiments has been conducted to investigate the effect of doping by carbon on the AlAs/GaAs SL disordering process.[12] Carbon in GaAs and AlAs is an acceptor species occupying the As site, $\mathbb{C}_{As}^{-}$. It has been found that, under As-rich (P_{As_4}=1 atm) annealing conditions, Eq. (3) together with the fact that $C_{I_{Ga}^{2+}}^{eq} \propto p^2$ have yielded calculated Al-Ga interdiffusivity values slightly higher than those obtained experimentally from the AlAs/GaAs SL disordering rate measurements. Under Ga-rich annealing conditions, however, the experimental Al-Ga interdiffusion data are lower than that predicted by the use of Eq. (4) by about 2 orders of magnitude. This is understood to be due to an undersaturation in I_{Ga}^{2+}, since such an I_{Ga}^{2+} undersaturation is present at the onset of experiments and the process of populating the SL via I_{Ga}^{2+} indiffusion from the sample surface is not effective.

For $\mathbb{C}_{As}^{-}$ itself, phenomena involve both $\mathbb{C}_{As}^{-}$ diffusion and the hole concentration reduction have been observed and analyzed. The experimental results, in consistency with analytical results, showed that carbon in GaAs is most likely an interstitial-substitutional species diffusing via the kick-out mechanism, and that the hole reduction is due to a precipitation process of the $\mathbb{C}_{As}^{-}$ atoms.[12]

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IV. PUBLICATIONS

A total of 14 journal papers, and 10 conference proceeding papers have been published, listed in the following.

IV.1. Archival journal papers

1. S. Yu, T. Y. Tan, and U. M. Gösele, "Diffusion Mechanism of Zinc and Beryllium in Gallium Arsenide," J. Appl. Phys. 69, 3547 (1991).

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IV.2. Conference proceeding papers

1. S. Yu, U. M. Gösele, and T. Y. Tan, "An Examination of the Mechanisms of Si Diffusion in GaAs," in "Impurities, Defects and Diffusion in Semiconductors: Bulk and Layered Structures," eds. D. J. Wolford, J. Bernholc, and E. E. Haller, Mat. Res. Soc. Symp. Proc. 163 (Mat. Res. Soc., Pittsburg, 1990) p.671.

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